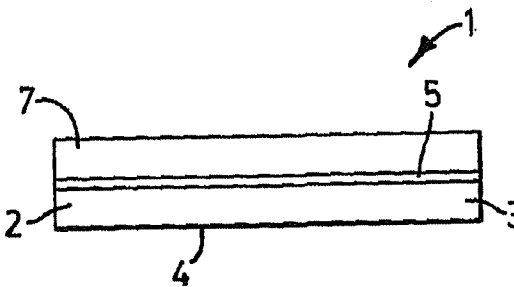




## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<b>(51) International Patent Classification <sup>6</sup> :</b> <b>B29C 47/02, 47/06, B32B 27/30, 31/30</b>	<b>A1</b>	<b>(11) International Publication Number:</b> <b>WO 98/52737</b> <b>(43) International Publication Date:</b> 26 November 1998 (26.11.98)
<b>(21) International Application Number:</b> PCT/CA98/00507 <b>(22) International Filing Date:</b> 25 May 1998 (25.05.98)  <b>(30) Priority Data:</b> 08/862,487           23 May 1997 (23.05.97)       US 08/863,596           27 May 1997 (27.05.97)       US  <b>(71)(72) Applicant and Inventor:</b> PLANETA, Mirek [CA/CA]; 45 Kingsbridge Garden Circle, Mississauga, Ontario L5R 3K4 (CA).  <b>(74) Agents:</b> MOSS, W., Dennis et al.; Barrigar & Moss, Suite 901, 2 Robert Speck Parkway, Mississauga, Ontario L4Z 1H8 (CA).		<b>(81) Designated States:</b> AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, GH, GM, GW, HU, ID, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report.</i> <i>Before the expiration of the time limit for amending the          claims and to be republished in the event of the receipt of          amendments.</i>
<b>(54) Title:</b> PVDC EXTRUSION METHOD AND PRODUCT		
<b>(57) Abstract</b>		
<p>A sheet material (1) for use in packaging substances requiring a stable barrier layer and a method for producing same are provided. The packaging material comprises a substrate sheet (3) coated with a layer of polyvinylidene chloride copolymer (7) having a thickness of at least 10 micrometers. Preferably, a thin layer of primer material (5) is located between the polyvinylidene chloride copolymer layer (7) and the substrate (3). The sheet material is prepared by extruding a single layer of polyvinylidene chloride copolymer (7) onto a moving sheet of the substrate (3). A standard coat hanger die which has been modified to provide shortened flow paths for the polyvinylidene chloride copolymer is utilized for the extrusion process. Prior to introducing the polyvinylidene chloride copolymer into the die, the inner surfaces of the flow paths in the die are coated with polyethylene in order to minimize adherence of polyvinylidene chloride copolymer to the inner surfaces of the flow paths.</p>		



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TITLE OF THE INVENTION

## PVDC EXTRUSION METHOD AND PRODUCT

BACKGROUND OF THE INVENTION

5           The present invention relates to generally sheet-like substrates which are extrusion coated with a layer of polyvinylidene chloride and its copolymers, and also a method for making polyvinylidene chloride extrusion coated materials.

10           The term PVDC hereinafter refers to copolymers including polyvinylidene chloride as one of the polymers. PVDC is a particularly effective material to be used as a barrier against moisture, oxygen and aromatic hydrocarbons. It is therefore desirable to use PVDC as a coating for packaging materials. The production of packaging materials typically involves coating a suitable substrate such as paper, PVC plastic or aluminum foil with PVDC, through a process of  
15           extrusion, to form a sheet material useful for packaging purposes. Applications for such packaging sheets include the formation of blister packs for the packaging of pharmaceuticals and meat and fish products. Also barrier coated substrates may be used to package substances such as milk, edible oils, nuts, cheese, spices and biscuits. Packaging sheets having a barrier layer of PVDC are effective for use  
20           with any product which requires protection from moisture, oxygen or which needs to retain gases, moisture or aroma.

          It is well known that it is difficult to extrude PVDC because of the susceptibility of PVDC to thermal degradation during extrusion. This is because  
25           of the very low temperature stability and autocatalytic degradation of PVDC. This degradation is accelerated by contact with metal ions from the die and extruder. Attempts have been made to overcome this problem by encapsulating the degradable PVDC material with non-degradable plastic material prior to extrusion. However, it is also well known that some applications require extrusion of PVDC

- 2 -

in a non-encapsulated form. Previous attempts to extrude PVDC in a non-encapsulated from a flat coat hanger die form have proved to be unsuccessful due to decomposition of the PVDC resulting from prolonged residence time of the PVDC in the die.

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Due to the inherent difficulties in extruding unencapsulated PVDC onto substrates, methods have been developed to coat substrates with liquid dispersions of PVDC latex as opposed extruding PVDC. One such method is known as the Gravura process. This is an expensive method which requires multiple passes of the substrate through the coating process. Each pass through the process corresponds to a single layer of dispersion being added to the substrate. This process often involves eighteen layers or more to be added to the substrate in order to provide a sheet with acceptable barrier properties. With thicker layers using this process, it is difficult to dry individual layers properly. This leads to the formation of pores in the coating thereby leading to porosity of the layers and reducing barrier properties. Thicker Gravura coatings are susceptible to flex cracking. Also, storage and handling of the water based emulsion used in the Gravura process is very difficult and time limited to approximately six months. These storage problems do not apply to PVDC extrusion resins.

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As mentioned above, difficulties in extruding PVDC plastics have resulted from the decomposition of PVDC into HCl and carbon during the extrusion process. The rate of decomposition is directly proportional to both the temperature at which the extrusion process is carried out and upon the length of time for the extrusion. Since the decomposition is autocatalytic, degradation of PVDC is rapid once the reaction has commenced.

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There therefore is a need for the provision of a packaging sheet material which is extrusion coated with a single unencapsulated layer of PVDC plastic.

- 3 -

There is a further need for a process for producing extrusion coated packaging material having a PVDC layer through a process of extrusion which can be carried out at lower temperatures and which involves a short residence time of the PVDC in the extruder and die. There is a need for such a process which makes use of a die formed of a material which does not catalyse the formation of hydrochloric acid from PVDC.

### SUMMARY OF THE INVENTION

The present invention is a sheet material comprising at least one layer of PVDC plastic extruded onto a generally planar substrate. The present invention further includes a method for extruding a single layer or multiple layers of PVDC plastic onto the substrate wherein the method is carried out at a relatively low temperature and with a small residence time of the PVDC in the extruder and die combination.

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According to one aspect of the invention there is provided an extrusion coated multilayer sheet material comprising a substrate layer, and a single non-encapsulated layer of polyvinylidene chloride copolymer having a thickness of at least 10 micrometers extrusion coated onto a surface of the substrate layer. The sheet material preferably includes an adhesive primer layer located between the substrate layer and the polyvinylidene chloride copolymer layer for facilitating bonding of the polyvinylidene chloride copolymer layer to the substrate layer.

According to another aspect of the invention, there is provided a method for coating a substrate sheet with a layer of polyvinylidene chloride comprising the steps of providing an extruder coupled to a die having a die opening, the die having shortened flow passages, coating the flow passages with polyethylene, passing molten polyvinylidene chloride from the extruder to the die and through the die opening, providing a primer means over a surface of the substrate,

transferring the substrate into close proximity to the die opening, and coating the substrate with the extruded polyvinylidene chloride emanating from the die opening.

5           According to still a further aspect of the invention, there is provided a method for extrusion coating a substrate sheet with a polyvinylidene chloride copolymer layer comprising the steps of providing an extrusion die for co-extruding in sheet form through a slot opening a partially encapsulated material comprising polyvinylidene chloride copolymer with an adhesive material bonded  
10 to one surface thereof, passing the substrate sheet by the slot opening; and extruding the partially encapsulated sheet material onto the substrate sheet so that the adhesive material is located between the substrate sheet and the polyvinylidene chloride. Preferably, the polyvinylidene chloride copolymer is by weight 80% to 99% polyvinylidene chloride and the substrate material is selected from the group  
15 consisting of polyvinyl chloride, polypropylene, polyester, polyamides, polystyrene, polyethylene, ethyl vinyl alcohol, polycarbonates, co-polymers and extrusion combinations thereof, paper and aluminum foil.

Further features and advantages will become apparent from the following  
20 detailed description taken in conjunction with the following drawings.

#### BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 is a sectional view of a preferred embodiment of PVDC coated sheet material according to the present invention;

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Figure 2 is a schematic diagram of the method of manufacturing the sheet material of the preferred embodiment;

Figure 3 is a perspective view of a die utilized in said method;

- 5 -

Figure 4 is a cross-sectional view taken along the lines IV-IV of Figure 3;

Figure 5 is a sectional view of an encapsulated embodiment of the present invention;

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Figure 6 is a sectional view of a partially encapsulated embodiment of the present invention;

Figure 7 is a schematic drawing showing a method of manufacturing the encapsulated embodiment of the sheet material of the present invention;

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Figure 8 is a schematic drawing showing a method of manufacturing the partially encapsulated embodiment of the sheet material of the present invention; and

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Figure 9 is a schematic drawing showing a further method of manufacturing the sheet material with hot melt adhesive as a primer; and

Figure 10 is a diagrammatic plan view of an extrusion die for producing partially encapsulated PVDC.

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#### DESCRIPTION OF THE PREFERRED EMBODIMENTS

As depicted in Figure 1, sheet material 1 is generally planar. The sheet material includes a substrate 3 having a top surface 2 and a bottom surface 4. A very thin coating of primer 5 is adhered to the top surface 2 of the substrate 3. The primer can include any combination of water or solvent soluble co-polymers or two component systems. The primer coating functions to improve the adherence of a layer of extruded PVDC 7 to the top surface 2 of the substrate 3. This allows

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- 6 -

the extrusion process to be carried out at a lower temperature while still achieving acceptable adherence of the PVDC to the substrate.

In an alternate embodiment of the present invention, the well known techniques of corona treatment and ozone treatment may be applied to either the top surface 2 or the bottom surface 4 of the substrate 3, or both, in order to improve the adherence qualities of these surfaces to the extruded PVDC 7. In this embodiment, the treated surfaces provide primer means for improving the adherence of the substrate to the layer of extruded PVDC 7.

10

The layer of PVDC 7 is applied to the top surface 2 of the substrate 3 so that the primer 5 is located between the PVDC layer 7 and the top surface 2 of the substrate 3. The fact that the PVDC layer is not encapsulated is an important feature of the sheet material 1. The PVDC layer 7 provides an excellent inert barrier against oxygen, moisture, oils and aromatics due to its highly crystalline structure. Oils and other additives from the packaged product cannot penetrate into the PVDC crystal in order to degrade the seal provided by the PVDC layer. The result is a highly durable and reusable sheet material due to the barrier qualities of the PVDC.

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The PVDC layer that is adhered to the substrate is a single layer of non-encapsulated PVDC having a generally uniform thickness of between 10 micrometers and 200 micrometers. The PVDC layer is produced from PVDC resin which is melted in an extruder, as described in more detail below. For the purposes of the present invention, a layer of PVDC refers to a layer of PVDC which is obtained from melting PVDC resin and includes by-products of the extrusion process which may become embedded in the PVDC. Furthermore, PVDC can also include usual additives such as stabilizers, plasticizers and processing aids. As used herein, PVDC refers to a co-polymer of polyvinylidene

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- 7 -

chloride in which polyvinylidene chloride preferably makes up 80% to 99% of the weight of the copolymer.

In an alternate embodiment both the top 2 and bottom 4 surfaces of the substrate 3 may be coated with a primer 5 and a layer of PVDC 7, as described above.

The substrate 3 may be chosen from a number of different substances such as PVC, polypropylene, polyester, polyamides, polystyrene, polyethylene, ethylene vinyl alcohol, polycarbonates and linear low density polyethylene (LLDPE). The substrate may also be formed of co-polymers of these substances and co-extrusion combinations may also be used as substrates. Paper and aluminum foil may also be used as substrates.

The method for producing sheet material 1 is shown schematically in Figure 2. Substrate 3 is provided on a roller 10. The substrate is then transferred mechanically according to any one of several methods known in the art from the roller to a series of work stations. Substrate 3 is first coated with a very thin layer of primer 5 at primer coat unit 12. The substrate 3 is then transferred to a drying unit 14 which serves to dry the coat of primer 5 which had previously been applied to the substrate 3. Once the primer coat has been dried sufficiently, the substrate 3 is brought into close proximity with a die opening 20 from which the extruded PVDC 7 is applied onto the moving substrate sheet. The final sheet material is then collected onto a collection roller 24.

25

As with known extrusion processes, PVDC resin is provided to extruder 16. The resin is then melted in the extruder and transferred to die 18. The molten PVDC is then extruded through die opening 20 so that a layer of PVDC 7 emanates from the die opening 20 for coating onto substrate 3, as described above.

- 8 -

A coat hanger die 18 similar to those known in the art is used for the purposes of the present invention. As shown in Figure 3, the die has a first body portion 30 and a second portion 32. The first and second body portions form a seam 36 at their junction when these are attached together. The die 18 includes a die entrance 28 and a die opening 20 through which the PVDC is extruded. As shown in Figure 4, the die also includes a generally parabolic inner cavity 38 having an inner surface 40.

The rate of the reaction involving the degradation of PVDC into HCl is proportional to the time in an exponential relationship. It is therefore necessary to limit the residence time of the molten PVDC in the die cavity to prevent this reaction from proceeding to any significant degree. This has been accomplished in the present invention by modifying a standard coat hanger die to shorten the flow passageways provided in the die cavity. For example, in the preferred embodiment, the distance between the die entrance 28 and the die opening 20 has been shortened to provide for a shortened flow path in comparison to commonly used coat hanger dies. Other methods of shortening the flow paths in the die which are known in the art are also possible for successfully carrying out the objectives of the present invention.

20

In order to prevent the degradation of PVDC from occurring in the die cavity 38, it is necessary to ensure that the rate of flow of PVDC at the peripheries of the die cavity is sufficient enough to avoid degradation. Previously, this has not been accomplished because the portion of PVDC adjacent to the inner die wall 40 would adhere to the die wall as the PVDC passes through the die cavity thereby impeding the movement of the portion of PVDC adjacent to the die wall. Also, PVDC tends to adhere to seam 36 thereby impeding its flow through the die. The result has been unequal flow rates of PVDC through the die cavity 38 with the PVDC at the centre flowing much more quickly than the PVDC at the peripheries

- 9 -

and at the seam 36. This leads to degradation of the slower flowing portions PVDC. Since the degradation of PVDC is autocatalytic in that the production of HCl catalyses the degradation, once degradation of PVDC begins at the peripheral portions, then this degradation will quickly spread to the central layers. In the present invention, the PVDC preferably passes from the extruder and through the die opening in approximately 5 minutes or less.

This problem has been solved by first coating the inner die walls 40 and the seam 36 between the first and second die members with a layer of polyethylene prior to extruding the PVDC. Since PVDC does not adhere to polyethylene, the body of PVDC at the peripheries of the die cavity now move through the die cavity 38 faster. This ensures the entire body of PVDC has substantially less residence time. As a result, the autocatalytic effect of the production of HCl has been significantly reduced.

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The use of a die made of a highly corrosion resistant metal, preferably Duranickel (trademark) or other high-nickel alloy steel, also inhibits the degradation of PVDC into HCl. Other materials or coatings can be used to slow down catalytic degradation of PVDC.

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In order to carry out the extrusion of PVDC successfully, it is very important to closely monitor the temperature of the PVDC as it is being melted. PVDC will rapidly degrade at elevated temperatures. Preferably, the extrusion of PVDC will be carried out at approximately 170°C. A cooling control package is used to maintain a tight (preferably  $\pm 2^\circ\text{C}$ ) temperature profile for the die and extrusion zones.

- 10 -

Once a layer or layers of PVDC have been successfully extruded onto a substrate, further materials can be extruded or co-extruded onto the substrate in order to improve the sealability of the sheet material.

5       An alternate embodiment of the present invention is shown in figure 5. Sheet material 1 is provided which includes a layer of extruded PVDC 7 which is fully encapsulated by a layer of adhesive material 8. The adhesive 8 is bonded to the substrate 3 along an outer side thereof. Similarly, the adhesive layer 8 is bonded to the PVDC layer along an inner side thereof.

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The adhesive layer may contain polyethylene and copolymers thereof, polypropylene and copolymers thereof including ethylene vinyl acetate, polyamides and copolymers thereof, polyesters and polyurethane and copolymers thereof. In addition, other adhesive materials known in the art such as hot melt  
15       adhesives may be used to form the adhesive layer. As known in the art, hot melt adhesives contain low molecular weight polymers having low temperature melting point. The adhesive layer 8 does not adhere to metal thereby rendering it unnecessary to coat the inner die walls with polyethylene. Furthermore, due to the lower viscosity of the adhesive layer 8, the encapsulated PVDC has a shorter  
20       residence time in the die and prevents the PVDC from touching the die walls. The adhesive layer also has better temperature stability.

The process for producing the fully encapsulated embodiment is set out in Figure 7. As is the case with the preferred embodiment, a sheet of substrate 3 is  
25       drawn from a roller 10. Since the adhesive layer 8 acts as a primer, it is unnecessary to add a primer to the sheet material prior to coating the substrate sheet 3. PVDC resin is fed into extruder 16 where the PVDC resin is melted. The melted PVDC is then delivered to feed block 45 where it is encapsulated by

- 11 -

adhesive in a manner known in the art. The encapsulated PVDC is then delivered to die 18 before being coated onto the substrate sheet 3 through die opening 20.

Figure 6 shows a partially encapsulated embodiment of an extrusion coated sheet material 50 of the present invention. A layer of PVDC 7 is bonded to an adhesive layer 8 which only partially encapsulates the PVDC layer 7. The adhesive layer 8 is bonded to substrate layer 3.

One preferred process for making the sheet material 50 is shown in Figure 8 (which is similar to the process of Figure 7, except that PVDC layer 7 is partially encapsulated instead of fully encapsulated). A sheet of substrate 3 is drawn from a feed roller (not shown). An extrusion die 54 is used to co-extrude the PVDC layer 7 and its partially encapsulating adhesive layer 8 onto substrate 3 in the vicinity of a nip between two rolls 56 and 58, thereby producing extrusion coated sheet material 50, which is rolled onto a take-up roller (not shown). The method for extruding partially encapsulated PVDC layer 7 of the sheet material 50 will now be described with reference to Figure 10 and Figure 6.

The method for extruding the partially encapsulated PVDC of sheet material 50 is based on the discovery that, when degradable plastic material is extruded from a slot die without encapsulation, most of the degradation occurs at the sides of the extrusion chamber rather than at its main surfaces, probably because of longer residence time at the sides.

Referring to Figures 6 and 10 the extrusion die 54 for producing partially encapsulated PVDC has a main body 112 with an extrusion chamber 114, a main inlet passage 116 for feeding a melt stream 118 of degradable plastic material, such as PVDC, into the extrusion chamber 114, and a slot 120 through which the plastic material is extruded in sheet form.

- 12 -

The die body 112 also has a pair of further inlet passages 124, 126 for feeding melt streams 128, 130 of non-degradable material (such as hot melt) to opposite side edges of the melt stream 118 of the PVDC. As shown in Figure 10, the melt streams 128, 130 of non-degradable plastic material prevent the degradable plastic material 18 from contacting the sides 132, 134 of the extrusion chamber 114. In addition to being fed to opposite sides of the melt stream, the non-degradable plastic material is also fed to one surface of the melt stream of the degradable plastic material to provide the main portion 7 of the plastic sheet extruded from the slot 120 with side portions 150 and 152 of non-degradable plastic material which also covers one face 156 of the main sheet portion 7, leaving the other surface 158 of main sheet portion 7 uncovered.

In practice, the die body 112 is usually formed of two parts secured together, one on top of the other, which produce a joint line extending around the sides of the extrusion chamber 114. The present invention thus keeps the degradable plastic material away from the joint line where it has been found most degradation of degradable plastic material is likely to occur.

The adhesive 8 of sheet material 50 is preferably a low temperature adhesive such as hot melt which has melt index higher than 1 MI and an adherence temperature range (hot tack range) of approximately 100° - 140° C. The low temperature adhesion range of hot melt allows the PVDC to be co-extruded at low temperatures, which minimizes degradation of the PVDC. The extrusion of the adhesive 8 with the PVDC layer 7 eliminates the need for the primer coating and drying step used in the process discussed above with reference to Figure 2.

An alternative method for making sheet material 50 is disclosed in Figure 9. The method of Figure 9 is similar to that described above with respect to Figure 8, however the adhesive layer 8 and PVDC layer 7 are independently extruded

- 13 -

onto substrate 3. In this regard, the adhesive layer 8 is first extruded onto the substrate 3 by an extrusion die 60, and then the PVDC layer 7 is extruded on top of the adhesive layer 8. The PVDC layer 7 is extruded in unencapsulated form using the method discussed above for extruding unencapsulated PVDC.

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Pressure sensitive adhesives could be used for adhesive layer 8 of material 50 in place of hot melt.

As will be apparent to those skilled in the art in the light of the foregoing  
10 disclosure, many alterations and modifications are possible in the practice of this invention without departing from the spirit or scope thereof. Accordingly, the scope of the invention is to be construed in accordance with the substance defined by the following claims.

## WHAT IS CLAIMED IS:

1. An extrusion coated multilayer sheet material comprising:  
a substrate layer; and  
a single non-encapsulated layer of polyvinylidene chloride copolymer having a thickness of at least 10 micrometers extrusion coated onto a surface of the substrate layer.
2. A sheet material according to claim 1 further including an adhesive primer layer located between the substrate layer and the polyvinylidene chloride copolymer layer for facilitating bonding of the polyvinylidene chloride copolymer layer to the substrate layer.
3. A sheet material according to claim 2 wherein the adhesive primer layer is comprised of a combination of water or solvent soluble copolymers, and in particular where such primer layer is a hot melt adhesive.
4. A sheet material according to claim 2 wherein the substrate layer is formed from a material selected from the group consisting of polyvinyl chloride; polypropylene; polyester; polyamides; polystyrene; polyethylene; ethylene vinyl alcohol; polycarbonates; copolymers, coextrusion combinations of the foregoing; paper and aluminum foil layer is formed from a sheet material selected from the group consisting of paper and aluminum foil.
5. A method for coating a substrate sheet with a layer of polyvinylidene chloride comprising the steps of:  
  
providing an extruder coupled to a die having a die opening, said die having shortened flow passages;



- 15 -

coating said flow passages with polyethylene;

passing molten polyvinylidene chloride from said extruder to said die and through said die opening;

providing a primer means over a surface of said substrate;

transferring said substrate into close proximity to said die opening; and

coating said substrate with said extruded polyvinylidene chloride emanating from said die opening.

6. A method according to claim 5 wherein the primer means includes a combination of water or solvent soluble copolymers.
7. A method according to claim 6 wherein said die opening is configured to yield a polyvinylidene chloride layer having thickness of at least 10 micrometers and said polyvinylidene chloride is extruded at a temperature of approximately 170° C..
8. A method according to claim 5 wherein said die and portions of said extruder in contact with the polyvinylidene chloride are comprised of a highly corrosion resistant compound, and in particular Duranickel..
9. A method for extrusion coating a substrate sheet with a polyvinylidene chloride copolymer layer comprising the steps of:

- 16 -

providing an extrusion die for co-extruding in sheet form through a slot opening a partially encapsulated material comprising polyvinylidene chloride copolymer with an adhesive material bonded to one surface thereof ;

passing the substrate sheet by the slot opening; and

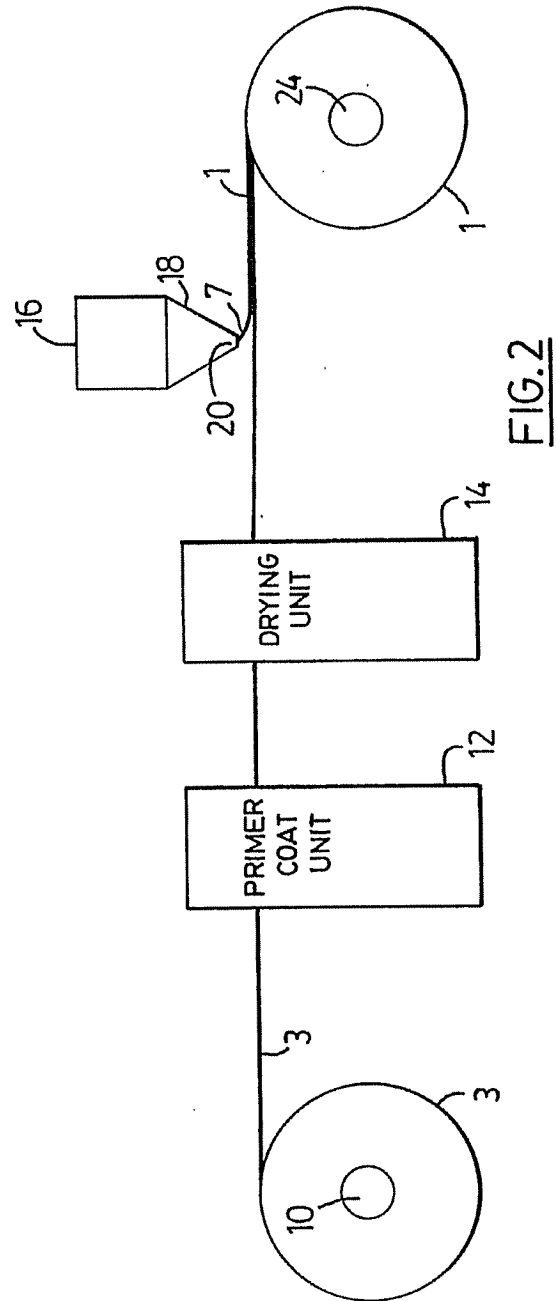
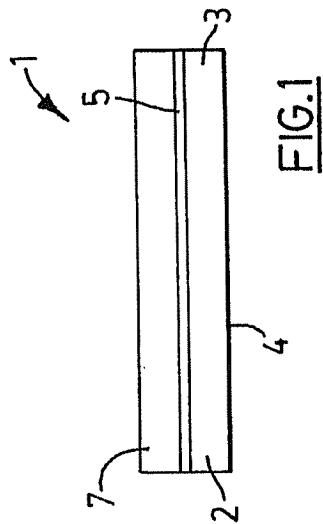
extruding the partially encapsulated sheet material onto the substrate sheet so that the adhesive material is located between the substrate sheet and the polyvinylidene chloride.

10. A method according to claim 9 wherein the adhesive material is a hot melt material having an MI greater than 1.

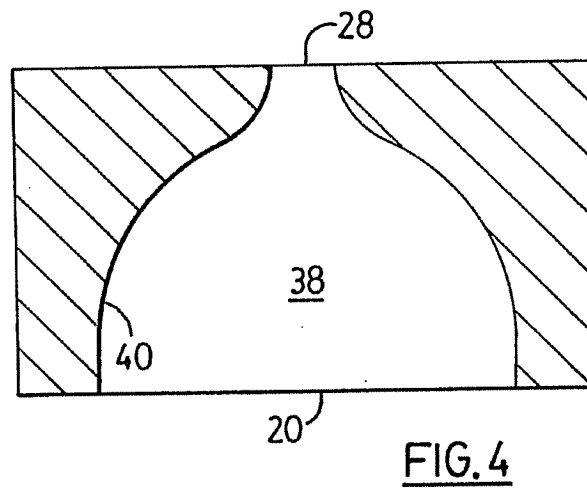
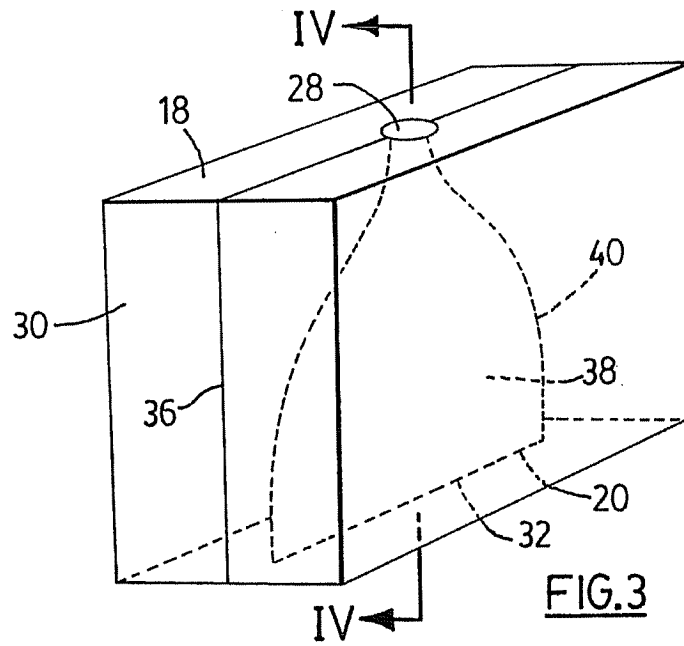
11. A method according to claim 9 wherein the polyvinylidene chloride copolymer is by weight 80% to 99% polyvinylidene chloride.

12. A method according to claim 9 wherein said substrate material is selected from the group consisting of polyvinyl chloride, polypropylene, polyester, polyamides, polystyrene, polyethylene, ethyl vinyl alcohol, polycarbonates, copolymers and extrusion combinations thereof, paper and aluminum foil.

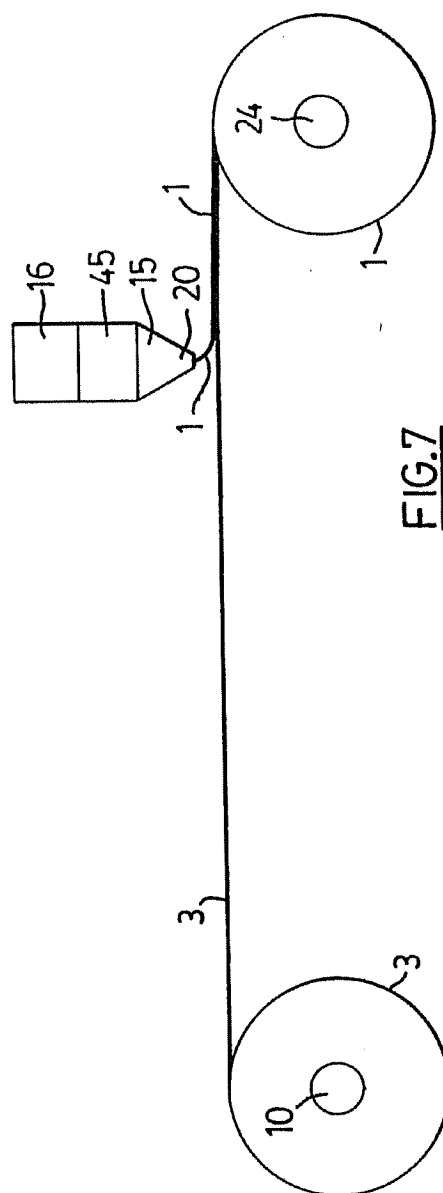
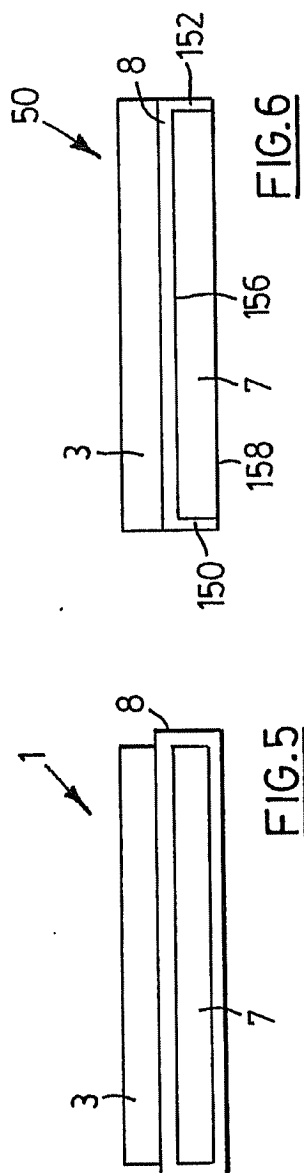
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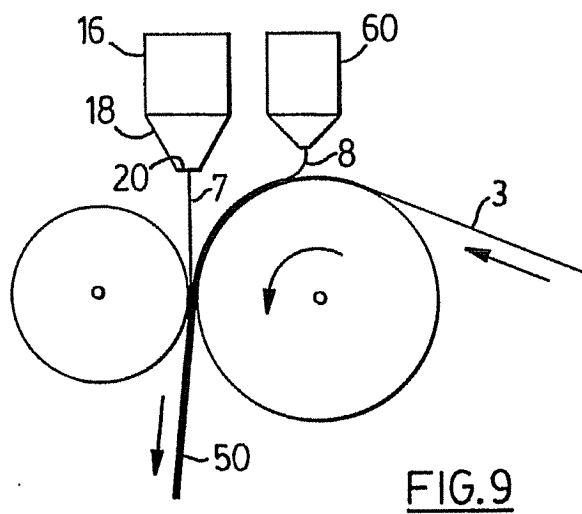
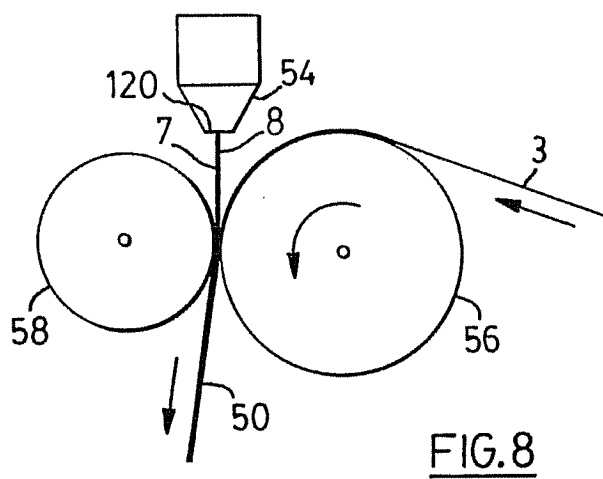
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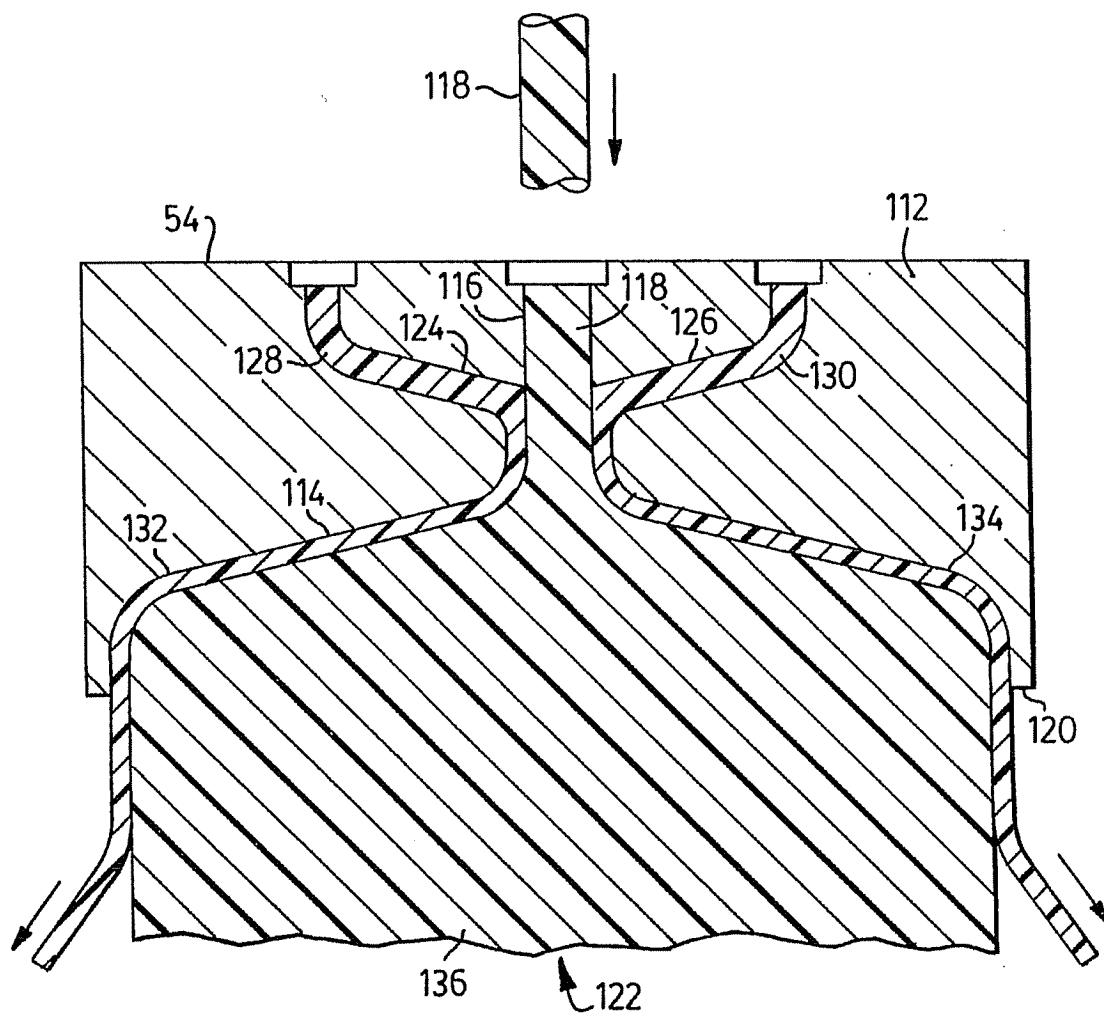


FIG.10

# INTERNATIONAL SEARCH REPORT

International Application No

PCT/CA 98/00507

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 B29C47/02 B29C47/06 B32B27/30 B32B31/30

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 B29C B32B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DATABASE WPI Section Ch, Week 9119 Derwent Publications Ltd., London, GB; Class A23, AN 91-137236 XP002076685 & JP 03 075 120 A (TOPPAN PRINTING CO LTD) see abstract	1-4
Y	---	5-12
Y	US 5 281 381 A (SU TIEN-KUEI) 25 January 1994 see the whole document ---	5-12
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☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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Date of the actual completion of the international search

7 September 1998

Date of mailing of the international search report

25/09/1998

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# INTERNATIONAL SEARCH REPORT

International Application No

PCT/CA 98/00507

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A	GB 1 142 606 A (THE DOW CHEMICAL COMPANY) 12 February 1969 see page 2, line 106 - line 118 see claims; figures ---	1,5-8
A	US 3 673 050 A (NEWMAN RITCHEY O JR ET AL) 27 June 1972 see the whole document -----	

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Information on patent family members

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